

## LANSCE DIVISION RESEARCH REVIEW

### ***Purified Plutonium Reduces Background Interference in Nuclear Cross-Section Measurements***

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#### **Introduction**

A program important for weapons diagnostics is currently under way to measure the  $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$  cross section. These cross-section measurements are performed with GEANIE (GERmanium Array for Neutron-Induced Excitations)<sup>1,2</sup> at the Weapons Neutron Research Facility<sup>3</sup> at Los Alamos National Laboratory by detecting gamma rays from the product nuclei. In early work, the relatively intense gamma-ray background from  $^{241}\text{Am}$  impurity in plutonium saturated the germanium detectors with background signals. Hence an improved plutonium sample purified of americium was fabricated.<sup>4</sup> This sample improved the  $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$  measurement. The same plutonium sample is being used in a new experiment to measure the  $^{239}\text{Pu}(n,n')^{239}\text{Pu}$  cross section—an important measurement for neutron transport calculations.

Plutonium enriched in  $^{239}\text{Pu}$  typically contains the isotopic impurities  $^{238}\text{Pu}$ ,  $^{240}\text{Pu}$ , and  $^{241}\text{Pu}$ . The  $^{241}\text{Pu}$  nuclei decay into  $^{241}\text{Am}$  by beta decay, and its concentration typically increases at a rate of 130 parts per million (ppm) per year, depending on the initial  $^{241}\text{Pu}$  concentration.<sup>5</sup> The gamma (  $\gamma$  ) rays emitted in  $^{241}\text{Am}$  decay are much more intense than those emitted from the longer-lived isotopes. Thus, the removal of  $^{241}\text{Am}$  provides an improved signal-to-background ratio for gamma-ray measurements. The purification technique and some measurements are described below.

#### **Sample Preparation**

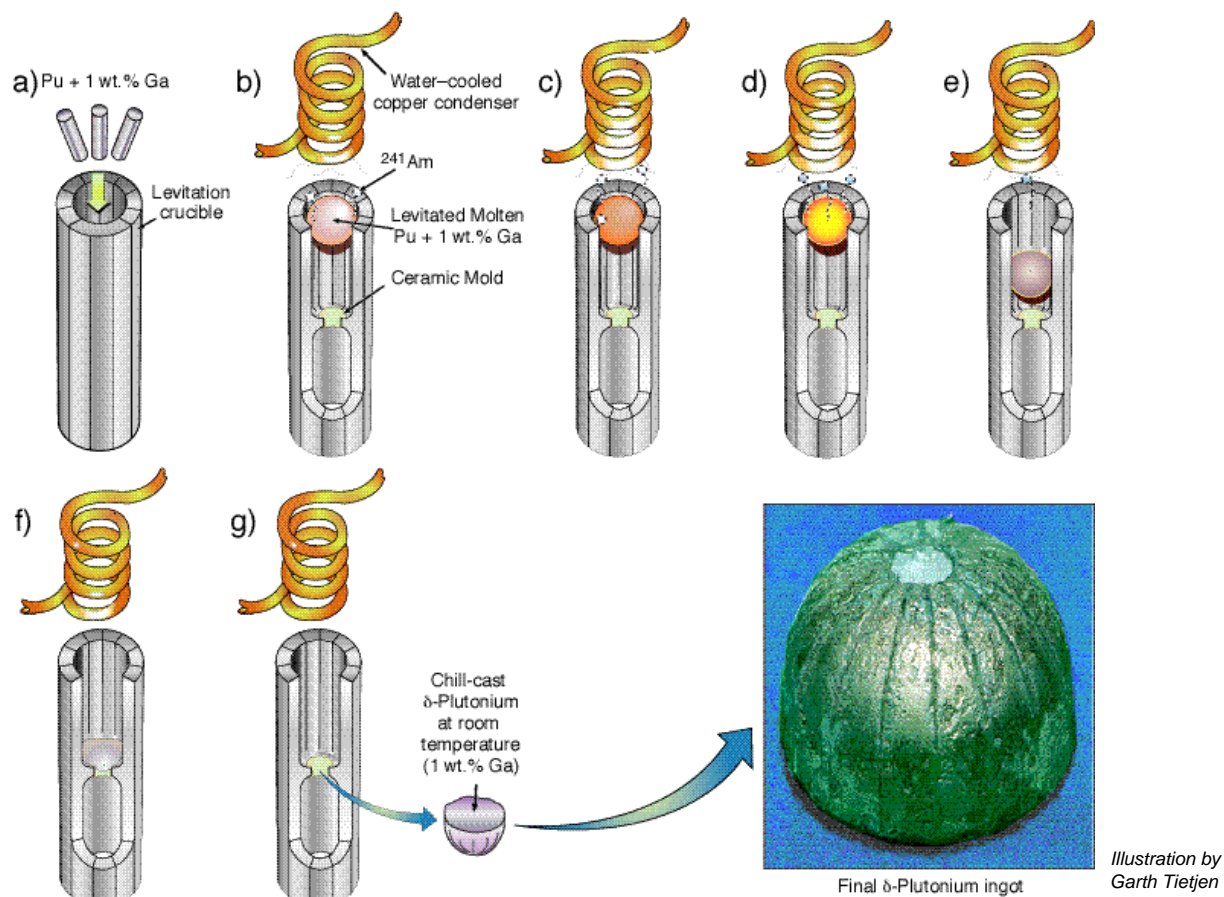
The plutonium sample used in the cross-section measurements on GEANIE was highly enriched  $^{239}\text{Pu}$ . This sample was processed through a molten-salt-extraction process followed by two electrorefining steps. The sample was then further purified through an electromagnetic-levitation distillation process. In this process, externally applied radio-frequency power operated at 4 kHz and 40 kW induces an electric current in the levitation crucible, which acts as a transformer inducing a current in the plutonium in the direction opposite to the current flowing in the crucible. An electromagnetic field is created,

and the plutonium sample in the crucible is repelled and levitated a small distance (2 to 3 mm) from the walls of the crucible. Approximately 200 g of material is processed in a single run. Because americium exhibits a high vapor pressure relative to plutonium,  $^{241}\text{Am}$  impurities, amounts typically greater than 100 ppm, are distilled away from the plutonium while in the melt and collected onto a water-cooled condenser. The material is distilled for 0.5 to 1 h. Enough gallium was added (1.0 wt %) to the plutonium during this purification process to stabilize the sample at the face-centered-cubic  $\gamma$ -phase upon solidification. As the power is reduced, the plutonium is chill-cast directly from the electromagnetic field into the crucible (Fig. 1).

A subsequent heat-treatment step produces a homogenous concentration of gallium throughout the plutonium sample. Gallium-stabilized  $\gamma$ -plutonium can be rolled to produce thin samples of uniform thickness. Thin (0.25 and 0.5 mm) sections (disks) of the purified plutonium sample were rolled, heat-treated, and encapsulated for safe handling at GEANIE. A small, thin sample is essential for the success of the gamma-ray technique used. In addition, the material used was selected for high  $^{239}\text{Pu}$  enrichment and low  $^{241}\text{Pu}$  content. Assays before and after the purification of the plutonium showed that the  $^{241}\text{Am}$  content was reduced by a factor of 200. The final  $^{241}\text{Am}$  content was 1.5 ppm.

#### **Nuclear Cross-Section Measurements**

The  $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$  cross section is important for weapons diagnostics, but knowledge of this cross section as a function of neutron energy is poor. A joint Los Alamos National Laboratory and Lawrence Livermore National Laboratory effort was initiated to measure this cross section using an indirect technique to determine partial  $\gamma$ -ray cross sections. To deduce the  $(n,2n)$  reaction-channel cross section, we used nuclear-reaction modeling to determine that portion of the cross section (weak  $\gamma$ -rays and internally converted  $\gamma$ -rays) that is not measured with our technique. The accuracy of the technique in part is dependent upon observing  $\gamma$ -ray transitions between low-lying states in the product nucleus.



▲ **Fig. 1.** Electrorefined plutonium is placed inside the levitation crucible along with sufficient gallium to stabilize the face-centered-cubic phase upon solidification (a). The mixture begins to melt and levitate a small distance from the crucible walls (b). The gallium begins to mix as the molten-charge plutonium and gallium is levitated, while at the same time volatile impurities like  $^{241}\text{Am}$  begin to distill away (c). The charge is allowed to remain in the melt to ensure uniform mixing (d). The furnace power is reduced, and the melt begins to solidify (e). While solidifying, the sample falls directly into a cold ceramic mold (the cooling rates are typically about 373 K per second) (f). An ingot of  $\delta$ -plutonium solidifies to conform to the shape of the crucible (g).

We use an array of 26 high-resolution germanium  $\gamma$ -ray detectors with background-suppression shields (GEANIE) in combination with the high-energy neutron beams from the Weapons Neutron Research (WNR) facility to measure nuclear-reaction cross sections. The excellent energy resolution of the GEANIE detectors allows us to identify specific reaction products from the characteristic  $\gamma$ -ray energies of low-lying nuclear excited states. The WNR neutron source provides a broad spectrum of incident neutron energies. We use time-of-flight techniques to determine the incident neutron energy for any particular  $\gamma$ -ray observed. This information gives us the probability or "cross section" for a specific nuclear reaction as a function of the incident neutron energy. Knowledge of these cross sections is useful in applications such as the calculation of integral quantities from microscopic modeling.

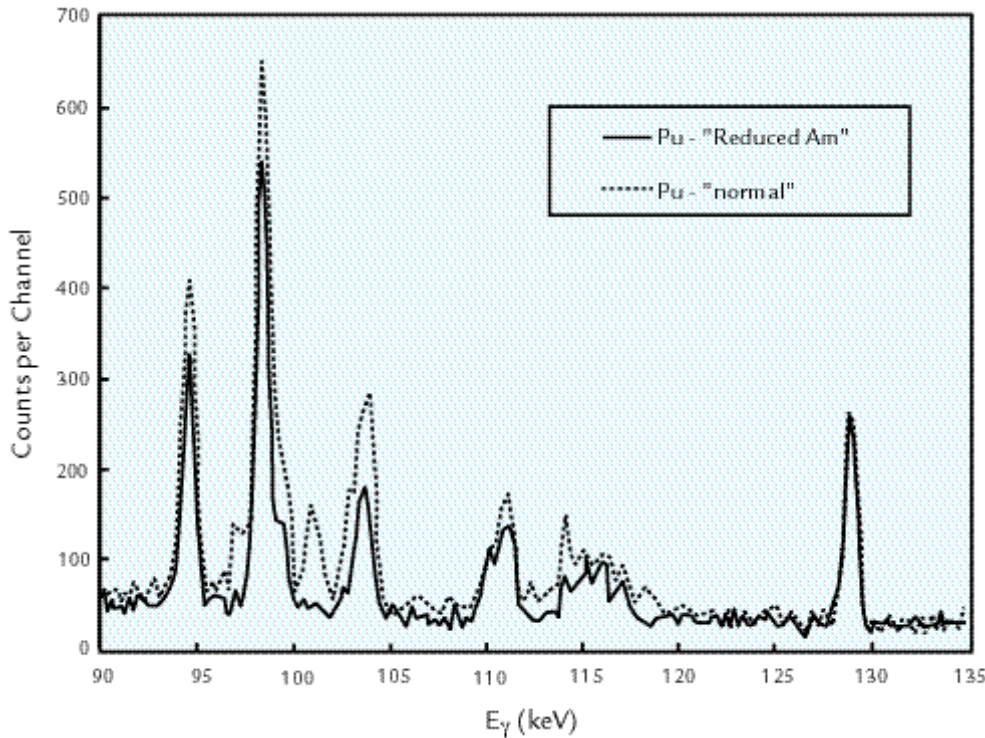
In these measurements, we want  $\gamma$ -rays from reactions on  $^{239}\text{Pu}$  to be the dominant signal; therefore, the enrichment in  $^{239}\text{Pu}$  must be as high as possible.

Our first measurements using a plutonium sample enriched to 94% in  $^{239}\text{Pu}$  indicated that the  $4^+ \rightarrow 2^+$  transition at 102 keV was obscured by a peak from the decay of  $^{241}\text{Am}$  in the sample. Removal of the  $^{241}\text{Am}$  greatly reduced the background interference in the GEANIE detectors (Fig. 2). But despite the reduction in background interference from  $^{241}\text{Am}$ , a strong contaminant line from fission still obscured the  $4^+ \rightarrow 2^+$  transition that we had hoped to observe. We are preparing a report on the results of this work.<sup>6</sup>

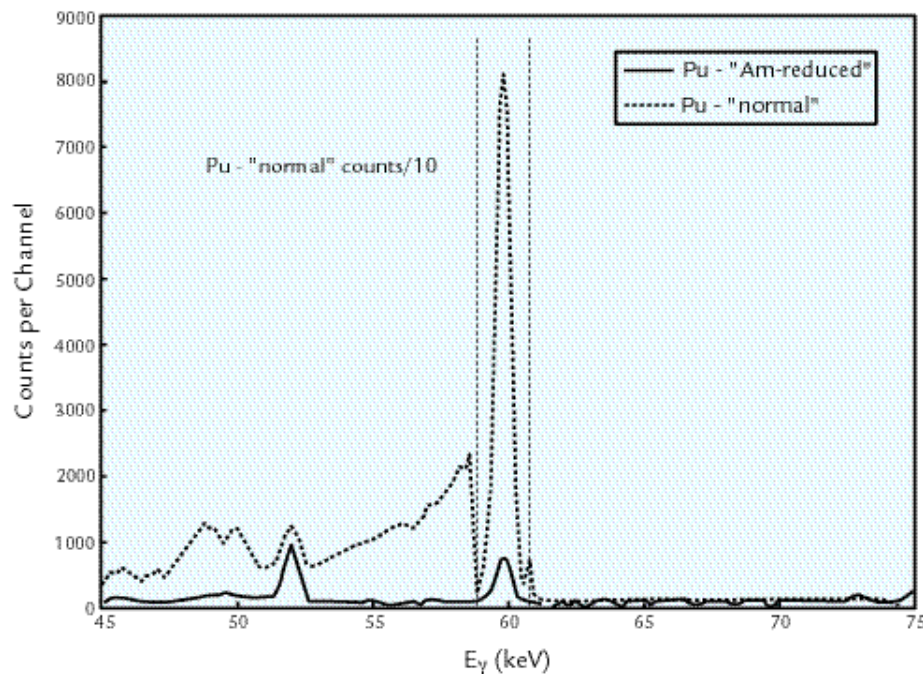
The use of the particular plutonium selected for recycling and purification also improved the measurement through a reduction in the amount of  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$  in the material.  $^{240}\text{Pu}(n,3n)^{238}\text{Pu}$  contributes background interference to the measurement at incident neutron energies greater than 12 MeV. Reduced  $^{241}\text{Pu}$  content in the sample selected for purification means that  $^{241}\text{Am}$  will be less of a problem when the sample is used in future measurements because less americium will grow into the sample.

The inelastic scattering of neutrons from plutonium is also an important quantity for calculations. This quantity, which is poorly known, is needed for neutron transport calculations. We are currently measuring the  $^{239}\text{Pu}(n,n')$  cross section to low-lying states using the same purified plutonium sample used in the  $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$  measurement, and we expect to observe thirty  $\gamma$ -ray lines from neutron inelastic scattering on  $^{239}\text{Pu}$ . Ten of these  $\gamma$ -ray lines overlap

in energy with  $\gamma$ -rays from  $^{241}\text{Am}$  decay. Reducing the  $^{241}\text{Am}$  in the plutonium sample is important for us to obtain more accurate inelastic scattering measurements. Fig. 3 shows the decrease in background from the very strong 59.5-keV line from  $^{241}\text{Am}$ . This greatly reduces the count rate in the detectors while allowing low-energy transitions to be observed more readily.



◀ **Fig. 2.** The plutonium sample background (i.e., no incident neutron beam) photon spectrum in the region of some of the plutonium and uranium K-shell x-rays (94 to 104 keV) is shown. The data were taken under identical conditions and normalized to match the "live time," which accounts for the acceptance of the data-acquisition electronics. A  $\gamma$ -ray from the decay of  $^{239}\text{Pu}$  is visible near  $E_\gamma = 129$  keV. A reduction in background near  $E_\gamma = 101.8$  keV was desirable for the  $\text{Pu}(n,2n)$  cross-section measurements and was achieved by reducing the  $^{241}\text{Am}$  content of the plutonium sample.



◀ **Fig. 3.** A background (i.e., no incident neutron beam), low-energy photon spectrum acquired with a "normal" plutonium sample containing  $^{241}\text{Am}$  "buildup" from the decay of  $^{241}\text{Pu}$  is plotted with a spectrum from an " $^{241}\text{Am}$ -reduced" plutonium sample that has undergone molten-salt extraction; zone refining; and the alloying, distillation, and chill-cast step. The intense 59.5-keV  $\gamma$ -ray from  $^{241}\text{Am}$  decay, which is near the center, demonstrates the huge reduction in counting rate achieved. The peak in the "normal" sample data has been scaled down by a factor of 10 to show the more detailed features in the spectrum. As in Fig. 2, the data were taken under identical conditions and normalized to match the live time. A  $\gamma$ -ray from the decay of  $^{239}\text{Pu}$  is visible near  $E_\gamma = 52$  keV.

## References

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